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Title: SEPARATOR FOR FUEL  
CELL AND  
MANUFACTURE  
METHOD FOR THE  
SAME

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UNDER 37 C.F.R. §1.10

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Derrick Brown

PRELIMINARY AMENDMENT

Commissioner for Patents  
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Washington, DC 20231

Sir:

Please amend the above-captioned application as follows:

***In the Specification:***

Please replace pages 1-16 of the specification with pages 1-17 of the enclosed substitute sheets. Applicant has also submitted herewith a strikethrough version of the specification indicating the amendments.

***In the Claims:***

Please cancel claims 1-3 without prejudice.

Please add the following claims.

4. (new) A separator for a fuel cell, comprising a molded body wherein the molded body is produced by filling a molding die with a graphite powder coated with a phenol resin and molding the graphite powder into a prescribed separator shape by applying pressure.
5. (new) The separator of claim 4, wherein the molded body comprises a resin content of about 10 wt.% to 24 wt.%, a bending strength of 40 MPa or higher, a gas permeability of  $10 \times 10^{-8}$  cc·cm/cm<sup>2</sup>·sec·atm or lower, and a volume resistance of  $10 \times 10^{-3} \Omega \cdot \text{cm}$  or lower.
6. (new) The separator of claim 4, wherein the molded body comprises a resin content of less than about 24 wt.%.
7. (new) The separator of claim 4, wherein the molded body comprises a resin content of about 14 wt.% to 18 wt.%.
8. (new) The separator of claim 4, wherein the graphite powder comprises graphite particles, and wherein gaps between the graphite particles are filled with the phenol resin.
9. (new) The separator of claim 4, wherein the separator comprises a thickness of about 1 mm to 3mm.
10. (new) A method for forming a separator for a fuel cell, comprising pressure-molding a resin-coated graphite powder into a prescribed separator shape at a molding pressure of 100 kg/cm<sup>2</sup> to

100 kg/cm<sup>2</sup> and a molding temperature of 120°C to 240°C, wherein the resin-coated graphite powder comprises an average particle diameter of 15 µm to 125 µm.

11. (new) The method of claim 10, wherein the separator has a resin content of less than about 24 wt.%.

12. (new) The method of claim 10, wherein the resin-coated graphite powder is formed by coating a graphite powder with a phenol resin.

13. (new) The method of claim 10, further comprising filling a molding die with the resin-coated graphite powder prior to pressure-molding the resin-coated graphite powder, wherein the molding die comprises the prescribed separator shape.

14. (new) The method of claim 10, further comprising forming the resin-coated graphite powder by mixing a graphite powder with a resin solution.

15. (new) The method of claim 10, wherein the resin-coated graphite powder comprises graphite particles, and wherein gaps between the graphite particles are substantially filled with the resin.

16. (new) A fuel cell, comprising:

one or more unit cells, wherein the one or more unit cells are stacked together, and wherein each unit cell, comprises:

an anode;

a cathode;

a solid polymer film, wherein the solid polymer film is sandwiched between the anode and the cathode; and

at least two separators, wherein the at least two separators sandwich the anode, the cathode, and the solid polymer film, and wherein each separator

comprises a resin content of less than about 24 wt.%.

17. (new) The fuel cell of claim 16, wherein each separator comprises a molded body, and wherein the molded body is produced by filling a molding die with a graphite powder coated with a phenol resin and molding the graphite powder into a prescribed separator shape by applying pressure.

18. (new) The fuel cell of claim 17, wherein the graphite powder comprises graphite particles, and wherein gaps between the graphite particles are filled with the phenol resin

19. (new) The fuel cell of claim 16, wherein at least one separator forms an oxidizing gas flow channel between the separator and a surface of the cathode.

20. (new) The fuel cell of claim 16, wherein at least one separator forms a fuel gas flow channel between the separator and a surface of the anode.

21. (new) The fuel cell of claim 16, wherein each unit cell comprises a cooling water flow channel.

22. (new) The fuel cell of claim 16, wherein each separator comprises a thickness of about 1 mm to 3mm.

23. (new) The fuel cell of claim 16, wherein each separator comprises a resin content between about 14 wt.% and 18 wt.%.

***In the Abstract:***

Please replace the abstract with the enclosed substitute sheet. Applicant has also submitted herewith a strikethrough version of the abstract indicating the amendments.

It is believed that no fees are due in connection with the filing of this Preliminary Amendment. However, if any fees are due, the Assistant Commissioner is hereby authorized to deduct said fees from Conley, Rose & Tayon Deposit Account No. 50-1505/5682-00300/EBM.

Respectfully submitted,



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~~SEPARATOR FOR FUEL CELL AND  
MANUFACTURE METHOD FOR THE SAME~~

~~TECHNICAL FIELD~~BACKGROUND OF THE INVENTION

1. FIELD OF THE INVENTION

The present invention relates to a separator for a fuel cell and to a ~~manufacture~~manufacturing method of the separator.

~~BACKGROUND OF THE INVENTION~~

2. DESCRIPTION OF THE RELATED ART

A fuel cell, ~~for example, (e.g., a solid polymer type)~~ is produced by ~~composing~~stacking unit cells. ~~each~~The unit cells are assembled by installing an anode and a cathode while sandwiching a solid polymer film between them. The unit cells are sandwiched between~~and~~ separators, ~~and b~~By stacking the unit cells in a number of several hundreds, a fuel cell is produced. A fuel gas such as hydrogen, or the like, is supplied through a gas supply groove formed in one separator in the anode side and an oxidizing gas such as oxygen, or the like, is supplied to the cathode side to cause an electrochemical reaction to convert the chemical energy ~~which of the fuel has into the electrical~~energy as output.

~~As a characteristic property as a material of the separators to be used for such a fuel cell,~~  
~~s~~Since electrical current generated in each unit cell flows through the separators and respectively neighboring unit cells are assembled as to compose a structure in series connection (in terms of a circuit) by closely attaching the separators of the respective unit cells to one another, the separators are required to have a contact resistance as low as possible between surfaces of the neighboring separators and between the contact surfaces of the separators and the electrodes closely attached to the separators. ~~and~~The separators are further required to have the an intrinsic resistance ~~of the separators themselves (hereinafter referred also as to~~ as "volume resistance") as low as possible ~~as well.~~

Further, since the fuel gas and the oxidizing gas are supplied to the respective electrodes while being completely separated from each other, a high degree of gas impermeability in a high degree is required. ~~Further, a~~ As described above, since a large number of unit cells are stuck together for assembly to be assembled, the thickness of the separators is made as thin as possible, ~~and e~~ Even if the separators are made thin as described, the separators are required to have sufficiently high mechanical strength and excellent molding precision as well from a viewpoint that fuel cells are assembled by stacking several hundreds of separators, then ~~and~~ fastening and affixing them together.

~~As separators required to have such characteristic properties, Some~~ well-known ones separators that have such characteristic properties can be made of ~~are isomg~~, for example, metal sheets ~~of such as pure copper, a stainless steel, and the like,~~ however, ~~i~~ In the case of such a metallic material, however, there ~~is~~ may be a problem ~~that with~~ material deterioration is ~~easy to~~ be caused by hydrogen embrittlement owing to from contact with hydrogen gas used as a the fuel gas, ~~and s~~ Such a metal materials ~~is~~ may be insufficient for ~~a long time~~ long-term stability.

Therefore, ~~these which~~ fuel cells that have recently been developed are ~~fuel cells~~ employing a molded body produced by mixing a graphite powder with a thermosetting resin ~~such as (e.g., phenol resin) as a binder and pressure~~ pressure-molding the resulting mixture ~~as into~~ separators. Since the graphite has a low electrical resistance and excellent corrosion resistance, the above described problems associated with using a metal separator are improved upon in the case of using a metal can be improved. Further, since the void gaps formed in the inside of the compacted powder molded body are filled with the binder, high gas impermeability, to a certain extent, can be obtained.

Such a separator made of graphite has conventionally been produced by, ~~for example,~~ using a resin-mixed graphite powder. The resin-mixed graphite powder is produced by steps of stirring thermosetting resin (such as a powder phenol resin) with a volatile organic solvent (such as an alcohol) to obtain a slurry, mixing and kneading a graphite powder with the slurry, drying the resulting mixture, and then pulverizing the dried mixture to a prescribed average particle

diameter. In the ~~above described~~ pulverization step, the graphite powder, whose surface is coated with the non-conductive resin by the kneading, is pulverized, and ~~owing to that~~ thus, ~~produced is a raw material powder of graphite whose surface is exposed~~ is produced. Then, ~~the~~ raw material powder is filled into a prescribed molding die and pressure-molded to form a separator for a fuel cell.

In this case, the resin content is higher, and the mechanical strength and the gas impermeability ~~become more excellent~~ are improved. Consequently ~~conventionally, a~~ separator made of graphite has been conventionally produced by specifying, at first, the resin amount sufficient to satisfy the factors such as the mechanical strength and the gas impermeability necessary for a separator of a fuel cell.

However, a conventional separator made of graphite produced by the above described production method does not necessarily satisfy the electrical characteristic properties required, such as volume resistance and the like. In other words, although the electrical characteristic properties become more excellent better as the resin amount is ~~less~~ reduced, the resin amount cannot be decreased so ~~much~~ significantly since the mechanical strength and the gas impermeability are decreased if as the resin amount is decreased, ~~and for that~~ Thus, a conventional separator made of graphite is may not provided with excellent electrical characteristic properties as well.

### SUMMARY OF THE INVENTION

~~The present invention is developed taking the above described problem into consideration and the purpose is to provide a~~ separator for a fuel cell may be made of a graphite and having have mechanical strength and gas impermeability as well as excellent electrical characteristic properties is described. ~~and to provide a~~ method for producing the such a separator is also described.

The separator for a fuel cell ~~of the present invention~~ is made of a molded body. The molded body is produced by filling a molding die with a graphite powder coated with resin on the surface and molding the powder by applying pressure into a prescribed separate shape.



That is, a conventional separator made of graphite is ~~may be produced by kneading resin and a graphite powder and then~~ obtaining a raw material powder containing graphite, whose The raw material powder surface is exposed by a pulverization step before the pressure-molding. ~~while taking~~ The conductivity after the molding may be taken into consideration and ~~pressure-molding for producing the raw material powder.~~ The separator is then produced by pressure-molding the raw material powder. ~~Consequently~~ In the process, the resin partially attaches to the surface of the graphite, and with a raw material powder in such a state, even if pressure-molding is carried out by a molding die filled with such a raw material powder, ~~the~~ an isostatic fluidity of respective particles through the resin cannot be obtained. That is, a graphite powder cannot sufficiently be coated with resin, ~~and for that~~ Thus, relatively large void gaps are easily left among the particles in a produced molded body. As a result, if the content of the resin is decreased, the strength and the gas impermeability are decreased.

On the other hand, ~~the~~ a separator of this invention is ~~may be produced by filling a molding die with a graphite powder whose surface is~~ has previously been sufficiently coated with resin, ~~and~~ Then, by pressure-molding the powder, ~~and in this case,~~ the fluidity of the respective graphite particles is improved in the molding die to produce a molded body with a small porosity. Further, the neighboring graphite particles are closely stuck to one another while ~~the resin being~~ voids are eliminated toward the voids among the graphite particles at the time of the pressure-molding, ~~and~~ e Consequently, the conductivity among graphite particles is ~~provided~~ also improved ~~at~~ after the pressure-molding.

~~As a result,~~ p Pressure molding of, for example, a graphite powder sufficiently coated with resin on the surface and having may have the a particle diameter of 15 to 125  $\mu\text{m}$  at the a molding pressure of 100 to 1,000  $\text{kg}/\text{cm}^2$  and the a molding temperature of 120 to 240°C. This process gives a molded body having 40 MPa of bending strength or higher and excellent in mechanical strength. ~~and~~ Gas permeability is 10 X-x 10<sup>-8</sup> cc·cm/cm<sup>2</sup>·sec·atm of gas permeability or lower and with a volume resistance of 10 X-x 10<sup>3</sup>  $\Omega$ ·cm of the volume resistance or lower and excellent in electrical characteristic properties even if the resin content is as low as 10 to 24 wt. %. ~~and~~ t The obtained molded body can suitably be used as a separator for a fuel cell.

## **BRIEF DESCRIPTION OF THE DRAWINGS**

Fig. 1 ~~is depicts~~ a schematic cross-sectional view showing the filling and pressurizing state of a resin-coated graphite powder in a molding die in the production process of a separator for a fuel cell ~~of the present invention~~;

Fig. 2 ~~is depicts~~ a schematic cross-sectional view showing the constitution of a fuel cell-based battery; and

Fig. 3 ~~is depicts~~ a front view of a separator incorporated in a unit cell.

## **DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS**

~~At first, the constitution and the operation~~ An embodiment of a solid polymer type fuel cell assembled using a separator ~~according to of the present invention will be is~~ described using a schematic figure of a general separator. As shown in Fig. 2, the fuel cell-based battery is assembled in a stack structure by stacking unit cells 5. Unit cells 5 may be stacked in a number of several hundreds. Each unit cell may be each composed of a solid polymer film 1, which is an ion exchange membrane made of fluoro resin, ~~for example, a~~ An anode 2 and a cathode 3 sandwiching may sandwich the solid polymer film 1 from both sides, ~~and s~~ Separators 4 may further sandwiching them from both sides; and ~~disposing dispose~~ current collector plates, which are omitted in the figure, in both sides of the united cells. Each anode 2 and each cathode 3 are made of carbon cloth woven from carbon fiber yarn, carbon paper, or carbon felt.

~~As showing shown~~ in Fig. 3, each separator 4 has fuel gas holes 6·7 and oxidizing gas holes 8·9 in the peripheral part to pass a hydrogen-containing fuel gas through and to pass an oxygen-containing oxidizing gas through, respectively. When the unit cells 5 are stacked, the respective holes 6 to 9 penetrate the inside of each fuel cell in the longitudinal direction to form a fuel gas supply manifold, a fuel gas discharge manifold, an oxidizing gas supply manifold, and an oxidizing gas discharge manifold.

In the inner side surface of the above described ~~each hole~~ holes 6 to 9, a flow channel of a groove part 12 with an optional pattern is formed. The pattern of the groove part 12 may be

formed to be, for example, a lattice like shape among a large number of projected parts, which may include shapes other than the shape illustrated in the figure. Through the groove part 12, as shown in Fig. 2, a fuel gas flow channel 13 is formed between the surface of an anode 2 and a separator 4 within the separator 4 in the anode 2 side, and a cooling water flow channel 14 is formed between the separator 4 and another neighboring separator 4. On the other hand, in a separator 4 in a cathode 3 side, an oxidizing gas flow channel 15 is formed between the separator 4 and the surface of the cathode 3.

In ~~such a~~ fuel cell-based battery with the above described structure, a hydrogen-containing fuel gas is supplied from a fuel gas supply apparatus installed ~~in-on~~ the outside to the fuel gas flow channel 13 of each unit cell 5 through the ~~above-described~~ fuel gas supply manifold, and in the anode 2 side of each unit cell 5, the electrochemical reaction,  $H_2 \rightarrow 2H^+ + 2e^-$ , is caused. The fuel gas, after the reaction, is discharged outside through the flue gas flow channel 13 of each unit cell 5 and the fuel gas discharge manifold.

At the same time, an oxygen-containing oxidizing gas (air) is supplied from an oxidizing gas supply apparatus installed ~~in-on~~ the outside to the oxidizing gas flow channel 15 of each unit cell 5 through the ~~above-described~~ oxidizing gas supply manifold, and in the anode 3 side of each unit cell 5, the electrochemical reaction,  $O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$ , is caused. The oxidizing gas, after the reaction, is discharged outside through the oxidizing gas flow channel 15 of each unit cell 5 and the oxidizing gas discharge manifold.

Following the above described electrochemical reactions, ~~as a whole~~ the electrochemical reaction,  $2H_2 + O_2 \rightarrow 2H_2O$ , proceeds, and by ~~the~~ this reaction, the chemical energy ~~which in~~ the fuel ~~has~~ is converted to the electrical energy and a prescribed battery function is performed. Incidentally, the fuel cell-based battery is operated in a desired temperature range of about 80 to 100°C, and during ~~the~~ operation, cooling water is supplied from a cooling water supply apparatus installed ~~in-on~~ the outside and circulated through the ~~above-described~~ cooling water channel 14 to keep the ~~operation~~ operating temperature within the ~~above-described~~ desired temperature range.

~~The said~~ Each separator 4 is generally formed ~~generally~~ to be a thin sheet-like shape with ~~the~~ a thickness of about 1 to 3 mm, ~~and a~~ A groove part 12 with ~~the~~ a depth of 0.3 to 1.5 mm is formed in both sides ~~in the case of a separator 4,~~ in the anode 2 side and ~~in one side in the case of a separator 4~~ in the cathode 3 side, so as to form the ~~said~~ fuel gas flow channel 13, the cooling water flow channel 14, and the oxidizing gas flow channel 15.

In an embodiment, ~~A~~ separator of the present invention ~~to be employed for a solid polymer type fuel cell-based battery just as described above~~ is made of a molded body produced by pressure-molding a graphite powder coated with resin on the surface into a prescribed separate shape under ~~the~~ molding conditions, which ~~will be~~ are described later. The molded body has excellent mechanical strength as ~~(e.g., 40 MPa of bending strength or higher), characteristic~~ excellent material propertyies as ~~a material of (e.g.,  $10 \times 10^{-8}$  cc·cm/cm<sup>2</sup>·sec·atm of gas permeability or lower), and~~ excellent electrical characteristic property properties of ~~(e.g.,  $10 \times 10^3 \Omega$ ·cm of the volume resistance or lower).~~ ~~although~~ ~~The content of resin in the molded body is about 10 to 24 wt.%, less than that of a conventional~~ ~~one~~ molded body. Hereinafter, the production method of such a separator made of graphite ~~will be~~ is described.

~~At first,~~ ~~The~~ graphite powder to be used may be any kind of graphite such as natural graphite, artificial graphite, carbon black, kish graphite, ~~and~~ expanded graphite, and the like, ~~and~~ The graphite powder may optionally be selected while taking ~~the~~ conditions of such as the cost into consideration. Natural graphite and artificial graphite ~~are preferable in terms of~~ ~~they~~ may be preferred due to their electrical properties. The average particle diameter of the graphite powder to be used ~~is preferable~~ may be in a range of 15 to 125  $\mu$ m. If ~~it~~ the average particle diameter is smaller than 15  $\mu$ m, the electrical resistance cannot ~~sufficiently~~ be decreased sufficiently and if it is larger than 125  $\mu$ m, the strength is deteriorated.

~~As the resin, most preferable is~~ A desirable phenol resin has excellent ~~in~~ wettability with the graphite powder. For example, ~~and also preferable are~~ any kind of resol type resin and novolak type resin may be used.

~~The A method for producing such a graphite powder coated with resin is not substantially limited. particularly restricted and, for example, a A resin solution may be diluted with an organic solvent with that has a low viscosity such (e.g., as methanol,).~~ The diluted resin solution may be, for example, a phenol resin solution. ~~The resin solution may be is-mixed with a graphite powder, and stirred and kneaded to-beinto a slurry. and-tThen the slurry is-may be granulated and dried by a spray drier to produce such-a graphite powder. In such a case, the mixing ratio at the above-described-mixing and kneading time is adjusted so as to control the resin content after the formation of the molded body to be 10 to 24 wt.%, preferably 14 to 18 wt.%. If the resin content is less than 10 wt.%, the-excellent gas impermeability is difficult to be obtained and if the content is more than 24 wt.%, the volume resistance and the contact resistance, which will-beare described later, are increased.~~

~~FurtherIn an embodiment,~~ the resin-coated graphite powder can be produced by a method in which the polymerization reaction of the resin takes place on the surface of the graphite powder during the stirring of the resin raw material solution. For example, in the case of forming a phenol resin coating, a graphite powder is further-added to a reaction container loaded with phenols, formaldehydes, a reaction catalyst, and another general reaction solvent. and-tThe mixture is heated to a prescribed temperature while being mixed and stirred to produce a resin-coated graphite powder. -in-which-tThe phenol resin adheres to the surface of the graphite powder and enters in lamellar graphite powder to firmly stick to the graphite powder.

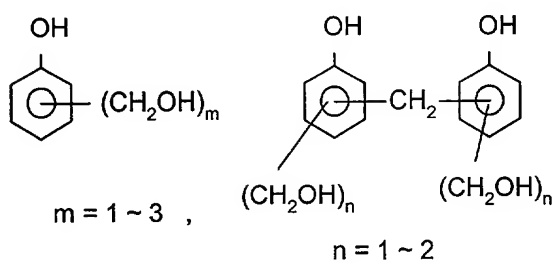
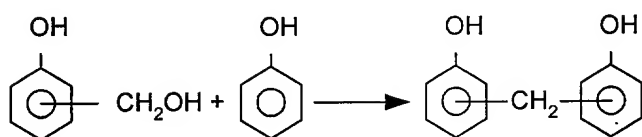
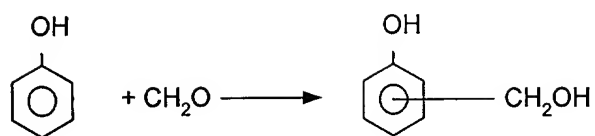
~~Incidentally, the said pPhenols denote-may include phenol and phenol derivatives. and eOther than phenol, examples are-include trifunctional ones-phenols such as m-cresol, resorcinol, and 3,5-xyleneol; tetrafunctional ones-phenols such as bisphenol A, and dihydroxydiphenylmethane; bifunctional o- or p-substituted phenols such as o-cresol, p-cresol, p-tert-butylphenol, p-phenylphenol, p-cumylphenol, p-nonylphenol, 2,4- or 2,6-xyleneol, and the like. FurtherIn some embodiments,~~ halophenols having chlorine or bromine as a substituent are also usable. and-other-than-using-solely-one-selected-from-those,-aA plurality of these-these compounds may also be used as-in a mixture.

~~As the f~~Formaldehydes, such as formalin may be used. ~~is most suitable and t~~Those in the form of paraformaldehyde may also be used. In some embodiments, ~~and besides,~~ some or most of the formaldehyde may be substituted with furfural or furfuryl alcohol ~~to be used.~~

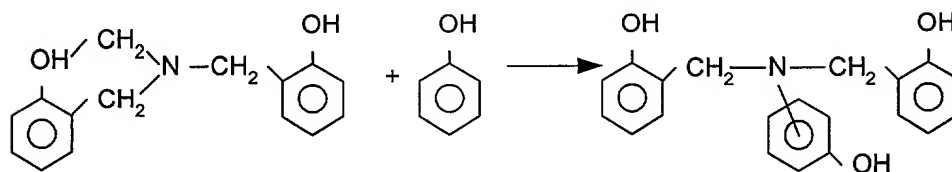
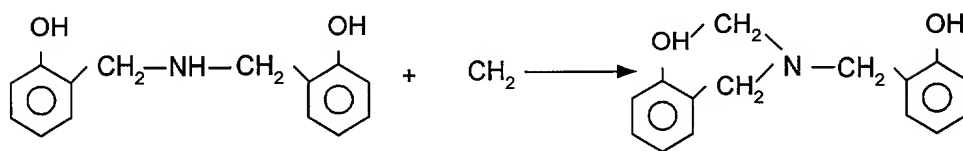
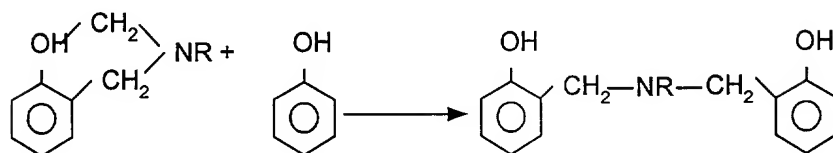
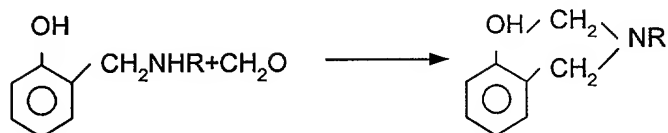
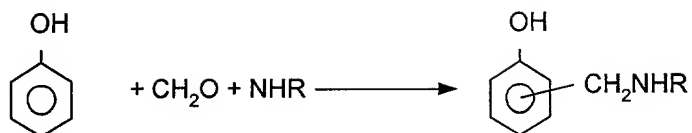
~~As the A~~ reaction catalyst, ~~preferable are~~ may include those capable of producing an -NCH<sub>2</sub>, ~~or~~ -OCH<sub>2</sub>, or -SCH<sub>2</sub> bond between the benzene ring of the phenols and a benzene ring as in a final structural formula of the phenol resin. For example, the reaction catalyst may include ~~usable are~~ hexamethylene tetramine, ammonia, and basic substances such as primary and secondary amines; (e.g., methylamine, dimethylamine, ethylenediamine, monoethanolamine and the like). ~~Further together with these, it is also possible to use~~ In some embodiments, a basic catalyst such as hydroxides of alkali metals and alkaline earth metals and tertiary amines may be used, which are commonly used at the time of phenol resin synthesis.

The following reaction formulas shown as Chem. 1 and Chem. 2 are examples of the general reaction formulas:

Chem. 1



Chem. 2



(in the formula, the reference character R denotes hydrogen or a lower alkyl such as methyl, ethyl and the like.).

In some embodiments, the phenol resin obtained is as shown for Chem. 1 and Chem. 2 may include a phenol resin having an -SCH<sub>2</sub> bond between the benzene rings other than the phenol resin shown as Chem. 1 and Chem. 2.

These phenols, formaldehydes, and the reaction catalysts are loaded into a reaction container such as a reaction vessel, and further a graphite powder is added and then the



reaction of the phenol and the formaldehydes is caused in the presence of the graphite powder. Consequently, a resin-coated graphite powder in which phenol resin firmly adheres to the surface of the graphite powder can be obtained.

The resin-coated graphite powder obtained by the above described production method is filled in a die having a molding space corresponding to a prescribed separator shape and pressure-molded. ~~in m~~ Molding conditions may include a molding pressure of 100 to 1,000 kg/cm<sup>2</sup> and a molding temperature of 120 to 240°C. If the molding pressure is lower than 100 kg/cm<sup>2</sup>, the density of the obtained molded body is low and the volume resistance ~~becomes~~ may be too high to make a separator with excellent ~~in~~ conductivity ~~hard to be obtained~~. On the other hand, if ~~excess plane~~ pressure higher than 1,000 kg/cm<sup>2</sup> is applied, the ~~phenomenon that resin is extruded out and the graphite particles and are unevenly distributed in the peripheral regions of the molded body, and thus dominantly takes place and for that the contact resistance, which will be described later,~~ is increased.

Although the molding temperature can properly be set corresponding to the heating properties of the resin, it ~~is~~ may be preferable to set the temperature generally at 120°C or higher ~~as described above in order to obtain the good fluidity of the graphite powder and the good molding property~~ properties in the molding die in ~~the case the resin content is small~~. If the temperature exceeds 240°C, ~~the a swelling phenomenon of in the molded body occurs, and if~~ the temperature is further increased, resin carbonization can takes place.

~~As described, t~~ The separator of the present invention, as shown schematically in Fig. 1, can be produced using a graphite powder 21 whose surface is coated with resin 20 by filling a molding die with the graphite powder and molding the graphite powder ~~in the above described~~ using molding conditions described herein. In the pressure-molding process, owing to the resin ~~softened~~ softening at first by heating, the respective graphite particles may show fluidity in responding to the resin in the molding die, ~~and due to that~~ Thus, the obtained molded body is ~~well~~ well-fitted with the die even after the resin is hardened, ~~that is i.e.,~~ the obtained molded body is provided with a high shape precision.

~~Further,~~ The resin 20 covering the surface of the respective raw material graphite particles 21 may flows towards the voids B among the respective raw material graphite particles 21 along the surface of the respective raw material graphite particles 21 in the regions A among the respective raw material graphite particles 21. The particles 21 may contacting one another during the molding pressure application. As a result, the surfaces of neighboring raw material graphite particles 21 are brought into contact with one another and a good electrical communication state is produced among these raw material graphite particles 21. Furthermore, the a resin extruded out in the regions where particles are brought into contact with one another as describe above may gathers in the gaps and be surrounded with the respective raw material graphite particles 21 to fill the gaps. In such a manner As such, fluidity of the resin 20 is evenly generated around the respective raw material graphite particles 21 entirely in on the inside of the molding die, so that the respective raw material graphite particles 21 surrounding the gaps are also firmly bonded to one another through the resin. Thus, filling the gaps to form a molded body.

~~In such a manner,~~ Even if the content of the resin is decreased, using raw material graphite particles whose surfaces are respectively coated with the resin makes it possible to obtain a molded body in which gaps formed among the respective raw material graphite particles are almost completely filled with the resin. and consequently, sSuch a molded body may have excellent in-mechanical strength and having a low gas permeability can be produced.

Moreover, under the above described pressure-molding conditions, since the resin is discharged towards the gaps among the raw material graphite particles 21 from neighboring regions of the raw material graphite particles 21 to bring these raw material graphite particles 21 into contact with one another, to obtain an excellent electrical communication state may be obtained. Thus, it is made possible to obtain a molded body that has an excellent also in the electrical characteristic property of the excellent volume resistance. For example, a volume resistance of  $10 \times 10^{-3} \Omega \cdot \text{cm}$  or lower may be attributed also to the low resin content as a whole.

~~Especially, as described above,~~ In the fuel cell-based battery composed by stacking unit cells 5 in a number of several hundreds, the inner resistance is considerably affected by the

contact resistance between mutually closely attached separators in neighboring unit cells 5-5 in addition to the volume resistance of the separator itself. In the case of the separator made of graphite ~~of the present invention~~ using a process as described herein, such a contact resistance is may be also extremely low. ~~That is~~ For example, in the case of a conventional separator made of graphite, the resin content cannot be decreased to, ~~for example~~, around 30 wt.% or lower because of the above described reasons. ~~and a~~ At the time when a graphite powder mixed with such a high amount of the resin is pressure-molded, the resin is extruded out to the peripheral part of the molded body from the gaps among the graphite particles to increase the resin ratio in the surface of the molded body. As a result, the separator has a high contact resistance as described above.

~~On the other hand~~ In an embodiment, in the case of the separator of the present invention, even if the resin content is decreased to 24 wt.% or lower, the desired mechanical strength and gas impermeability can be maintained, ~~and~~ The separator obtained is may be provided with a low contact resistance as described above. ~~Consequently,~~ Using such a separator, a fuel cell-based battery with low inner resistance and high power generation efficiency can be assembled. Further, the thickness of the separator can be made relatively thin and consequently, the fuel cell-based battery can be made small and light in weight.

### Examples

Hereinafter, the present invention will be described along with examples, however the present invention is not at all restricted to these examples or to the above described embodiments. ~~and~~ It is possible for the present invention to be modified to be variously modified within the true scope of the invention.

#### Example 1

A reaction container was loaded with a graphite powder (with ~~the an~~ an average particle size of 100  $\mu\text{m}$ ), phenol, formaldehyde, reaction catalyst (hexamethylenetetramine or ammonia together with a caustic soda solution), and a reaction solvent. ~~and w~~ While being mixed and stirred, the mixture was heated at 80°C for 1 hour. The content of the reaction container was cooled to a room temperature and after the stirring was stopped, the black granular substance

precipitated separately from the solvent in the reaction container was discharged out and washed with water. The resulting substance was ~~filtered to be separated from the solvent~~ by filtering and dried to obtain a resin-coated graphite powder. The content of the resin in the resin-coated graphite was 14%.

Then, the resin-coated graphite powder was filled into a molding die and pressure-molded at a molding pressure of 200 kg/cm<sup>2</sup> and a molding temperature of 160°C to produce a specimen for measuring a variety of characteristic properties, which ~~will be~~ are described later.

#### Example 2

A resin-coated graphite powder was produced in the same manner as ~~the~~ in example 1, except that ~~the~~ a raw material graphite powder with ~~the~~ an average particle diameter of 60 μm was used in place of the graphite powder in ~~the~~ example 1. The content of the resin in the resin-coated graphite was 18%. Next, using the obtained resin-coated graphite powder, a specimen was produced in the same molding manner as ~~described above~~ in the same molding conditions as those of ~~the~~ example 1.

#### Example 3

A resin-coated graphite powder was produced in the same manner as ~~the~~ in example 1, except that ~~the~~ a raw material graphite powder with ~~the~~ an average particle diameter of 45 μm was used in place of the graphite powder in ~~the~~ example 1. The content of the resin in the resin-coated graphite was 21%. Next, using the obtained resin-coated graphite powder, a specimen was produced in the same molding manner as ~~described above~~ in the same molding conditions as those of ~~the~~ example 1.

#### Comparative example 1

A resin-coated graphite powder was produced in the same manner as ~~the~~ in example 1, except that ~~the~~ a raw material graphite powder with ~~the~~ an average particle diameter of 25 μm was used in place of the graphite powder in ~~the~~ example 1. The content of the resin in the resin-

coated graphite was 25%. Next, using the obtained resin-coated graphite powder, a specimen was produced in the same molding manner as described above in the same molding conditions as those of the example 1.

#### Comparative example 2

A resin-coated graphite powder was produced in the same manner as the in example 1, except that the a raw material graphite powder with the an average particle diameter of 130  $\mu\text{m}$  was used in place of the graphite powder in the example 1. The content of the resin in the resin-coated graphite was 8%. Next, using the obtained resin-coated graphite powder, a specimen was produced in the same molding manner as described above in the same molding conditions as those of the example 1.

#### Comparative example 3

After the powdery phenol resin was pulverized and mixed by a ball mill, methanol was added to obtain a slurry of the phenol resin. and t The slurry was mixed with a raw material graphite powder with the an average particle diameter of 125  $\mu\text{m}$ , similar to that of the example 1. and a After being stirred at 60°C for drying, the resulting mixture was pulverized by a mixer to obtain a resin-graphite mixed powder. Additionally, magnesium stearate was added in the middle in of the pulverizing and mixing step of the phenol resin.

The content of the resin in the resin-graphite mixed powder obtained in the above described manner was 18% and using the obtained mixed powder, a specimen was produced in the same pressure-molding manner as described above in the same molding conditions as those of the example 1.

Using the respective specimens obtained in examples 1 to 3 and comparative examples 1 to 3, the compressive strength, the bending strength, the contact resistance, the intrinsic resistance, and the gas permeability were measured according to the following respective measurement methods. Results are shown in Table 1.

Measurement method:

(1) compressive strength

measured according to JIS K 7208 (specimen: 10 mm square ~~X~~x 4 mm height);

(2) bending strength

measured according to JIS K 7203 (specimen: 10 mm width ~~X~~x 4 mm height ~~X~~x 80 mm length);

(3) contact resistance

voltage was measured by laminating two specimens (20 mm square ~~X~~x 1 mm thickness) between measurement electrodes, applying and contact plane pressure of 25 kg/cm<sup>2</sup>, and applying electric current of 1 A and then contact resistance was calculated;

(4) intrinsic resistance

volume resistivity was measured according to JIS K 7194 (2 mm plate thickness); and

(5) gas permeability

gas permeation amount of nitrogen gas was measured while the pressure difference of 1 atm being generated between both sides of a specimen and the gas permeability was calculated.

Table 1

		graphite average particle diameter	resin content	compressive strength	bending strength	contact resistance	intrinsic resistance	gas permeability
		$\mu\text{m}$	%	MPa	MPa	$\Omega \text{ cm}^2$	$\Omega \text{ cm}$	$\frac{\text{cc} \cdot \text{cm}}{\text{cm}^2 \cdot \text{s} \cdot \text{atm}}$
example	1	100	14	62	44	$5 \times 10^{-3}$	$3 \times 10^{-3}$	not higher than $5 \times 10^{-6}$
	2	60	18	81	49	$8 \times 10^{-3}$	$6 \times 10^{-3}$	not higher than $5 \times 10^{-6}$
	3	45	21	94	53	$8 \times 10^{-3}$	$6 \times 10^{-3}$	not higher than $5 \times 10^{-6}$
comparative example	1	25	25	113	60	$15 \times 10^{-3}$	$11 \times 10^{-3}$	not higher than $5 \times 10^{-6}$
	2	130	8	50	30	$5 \times 10^{-3}$	$3 \times 10^{-3}$	$5 \times 10^{-6}$
	3	125	18	60	38	$10 \times 10^{-3}$	$8.7 \times 10^{-3}$	$5 \times 10^{-3}$

As ~~showing~~shown in Table 1, even if the resin-coated graphite powder is used, the contact resistance and the intrinsic resistance were high in the comparative example 1 where the resin content exceeds 24 wt.%, and ~~t~~The compressive strength and the bending strength were low and the gas permeability was high in the comparative example 2 where the resin content was less than 10 wt.%. ~~On the other hand, i~~In the comparative example 3, ~~where used in place of the resin-coated graphite powder was the~~ a resin-graphite mixed powder produced by adding a graphite powder to the resin, stirring them, and then pulverizing the resulting mixture was used

~~in place of the resin-coated graphite powder,~~ the bending strength was low and the specimen was inferior in gas permeability as well as the contact resistance and the intrinsic resistance.

~~Contrary, i~~In the examples 1 to 3 where resin-coated graphite powders ~~with the~~had an average particle diameter of 10 to 125  $\mu\text{m}$  and ~~the a~~a resin content of 10 to 24 wt.%, ~~the~~molded bodies obtained were excellent in ~~the~~compressive strength and bending strength and had low contact resistance and intrinsic resistance.

~~Incidentally~~Additionally, further specimens (1.5 mm thickness ~~X x~~250 mm length X x 250 mm width) for molding precision were produced and the thickness was measured at 25 points of a lattice. ~~and t~~The average value and the difference of the respective measured values from the average value were calculated to evaluate the molding precision. ~~and a~~Any specimen of ~~the~~examples 1 to 3 was found ~~the~~with a thickness precision within  $\pm 15 \mu\text{m}$  or lower, ~~also~~which showing shows excellent molding precision.

As described above, ~~since the a~~separator of the present invention produced by pressure-molding a graphite powder whose surface is coated with resin has excellent mechanical strength, ~~the~~gas impermeability, and ~~the~~excellent electrical characteristic properties, ~~a~~a fuel cell-based battery with excellent capability can be produced using ~~the~~such a molded body.



## ABSTRACT OF THE DISCLOSURE

To provide a separator for a fuel cell made of graphite may be excellent in mechanical strength and gas impermeability as well as electrical characteristic properties, a separator for a fuel cell is produced from a molded body that is produced by filling a graphite powder ~~21~~ whose surface is coated with resin ~~20~~ in a molding die and by pressure-molding the powder. Such a molded body is ~~provided~~ provides with excellent mechanical strength, ~~of 40 MPa of bending strength or higher,  $10 \times 10^{-8}$  cc·cm/cm<sup>2</sup>·sec·atm of gas permeability, or lower, and electrical property properties of  $10 \times 10^{-3}$   $\Omega$ ·cm of the volume resistance or lower~~ even if the content of resin is decreased to as low as 10 to 24 wt.%.